

NO_x Reduction on a transition metal-free γ -Al₂O₃ catalyst using Dimethylether (DME) as a reducing agent

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Introduction

Due to their considerable fuel efficiencies, diesel-engine vehicles form an increasingly large portion of the world's automotive market. On the other hand, the high air to fuel (A/F) ratios that are used in these engines prevent the application of traditional three-way catalysts (TWC) for the abatement of the hazardous pollutants such as NO_x that are emitted by these mobile sources. Catalytically there are two general pathways for NO_x reduction in an automotive catalyst [1]: direct NO_x decomposition on the catalyst or selective catalytic reduction (SCR). In order to tackle the problem of selective catalytic reduction of NO_x originating from lean-burn engines, use of short chain alcohols or ethers has recently received increasing attention [2-5]. In particular, it has been shown that methanol and dimethyl ether (DME) can be effectively used for NO_x reduction purposes under lean conditions [4-5].

Materials and Methods

For the experiments given in this work, a promoted, transition metal-free γ -Al₂O₃ catalyst was prepared. Structural and spectroscopic characterization of the catalyst was also performed. Catalytic behavior of the transition metal-free promoted γ -Al₂O₃ catalyst was investigated by DME adsorption, NO_x adsorption, methanol adsorption and various co-adsorption experiments where transmission infrared spectroscopy and mass spectroscopic methods were utilized as the major instrumental tools.

Results and Discussion

We have investigated interaction of NO₂ and DME with a modified γ -Al₂O₃ catalyst using in situ transmission infrared spectroscopy (figure 1a), residual gas analysis (RGA) (figure 1b) and temperature programmed desorption (TPD) techniques. Reduction of NO₂ with DME was studied in a batch reactor at various temperatures and at different relative partial pressures of reactants. These studies were also accompanied by similar experiments using isotopically labeled N¹⁵O₂. As methanol is one of the crucial surface species that is relevant to DME decomposition on alumina, methanol adsorption, decomposition and reaction with NO₂ and N¹⁵O₂ was also examined. Analogous experiments were also carried out using ¹³CH₃OH as a further support for infrared spectroscopic assignments. Nature and relative

quantities of the gas phase products were examined using gas phase IR spectroscopy, RGA and TPD techniques. Finally, the effect of the presence of additional H₂O or O₂ in the reactant mixture on the reaction mechanism and the type of the surface species observed during the reduction of NO_x with DME is discussed.

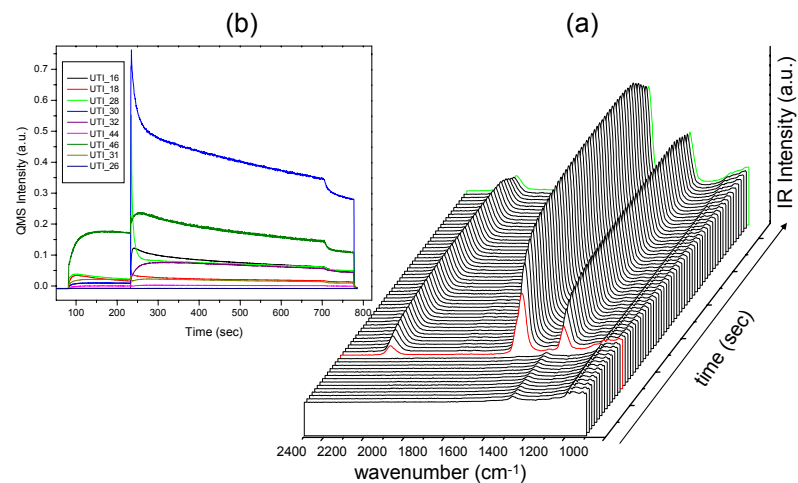


Figure 1. (a) Time resolved FTIR data (b) RGA data for the adsorption and reaction of DME with NO₂ on a transition metal-free promoted γ -Al₂O₃ catalyst at 573 K

Significance

In this study we investigated the catalytically relevant surface species that are formed during the adsorption of DME on a transition metal-free promoted γ -Al₂O₃ catalyst at various temperatures and also examined the behavior of surface species during the course of NO_x reduction as an attempt to link catalytic activity to the nature of surface species that are formed or present under catalytic reaction conditions.

References

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